Form PTO-1390 (Rev. 5-93) US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NO.
H 3763 PCT/US

U.S. APPLICATION NO. (if known, see 37 CFR 1.5)
09/868379

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/EP99/09683	**December 9, 1999**	**December 18, 1998**

TITLE OF INVENTION
FINE SUSPENSIONS OF POORLY SOLUBLE CALCIUM SALTS AND THEIR USE IN DENTAL CARE PRODUCTS

APPLICANT(S) FOR DO/EO/US
Christian KROPF, Ulrike BRUENINGHAUS, Amergio PASTURA, Michael MEINDERS, Peter WUELKNITZ, Rolf HEMPELMANN and Marcel ROTH

Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:

1. ■ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.

2. □ This a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.

3. □ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).

4. ■ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.

5. ■ A copy of the International Application as filed (35 U.S. C. 371(c)(2)).
   a. □ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ■ has been transmitted by the International Bureau.
   c. □ is not required, as the application was filed in the United States Receiving Office (RO/US).

6. ■ A translation of the International Application into English (35 U.S.C. 371(c)(2)).

7. ■ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. □ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. □ have been transmitted by the International Bureau.
   c. □ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ■ have not been made and will not be made.

8. □ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).

9. ■ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).

10. □ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11. to 16. below concern other document(s) or information included:

11. □ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.

12. □ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.

13. ■ A FIRST preliminary amendment
    □ A SECOND or SUBSEQUENT preliminary amendment.

14. □ A substitute specification.

15. □ A change of power of attorney and/or address letter.

16. □ Other items or information.:

"Express Mail" mailing label number EL 615775936 US

U.S. Application No. (if known see CFR1.30)	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
09/868379	PCT/EP99/09683	H 3763 PCT/US

17. ■ The following fees are submitted:

Basic National Fee (37 CFR 1.492(a)(1)-(5)):

Search Report has been prepared by the EPO or JPO..... **$860.00**

International preliminary examination fee paid to USPTO (37CFR 1.482) **$670.00**

No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37CFR 1.445(a)(2))..... **$690.00**

Neither international preliminary examination fee (37CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO..... **$970.00**

International preliminary examination fee paid to USPTO (37CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)..... **$96.00**

	CALCULATIONS	PTO USE ONLY
ENTER APPROPRIATE BASIC FEE AMOUNT =	$ 860	00
Surcharge of **$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date 37 (CFR 1.492(e)).	$ 0	00

Claims	Number filed	Number Extra	Rate		
Total Claims	7 - 20 =	0	0 X $18.00	$ 0	00
Independent Claims	4 - 3 =	0	0 X $80.00	$ 80	00
Multiple dependent claims (s)(if applicable)		0	+ $270.00	$ 0	00
TOTAL OF ABOVE CALCULATIONS =				$ 940	00
Reduction by ½ for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).				$ 0	00
SUBTOTAL =				$ 940	00
Processing fee of **$130.00** for furnishing the English translation later the ☐ 20 ☐ 30 months from the earliest claimed priority date (37CFR 1.492(f)). +				$ 0	00
TOTAL NATIONAL FEE =				$ 940	00
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). $40.00 per property +				$ 0	00
TOTAL FEES ENCLOSED =				$ 940	00
				Amount to be: refunded	$
				charged	$940.00

a. ☐ A check in the amount of $_____ to cover the above fees is enclosed.

b. ■ Please charge my Deposit Account No. 01-1250 in the amount of $940.00 to cover the above fees. A triplicate copy of this sheet is enclosed. Order No. 01-0461.

c. ■ The Assistant Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 01-1250. A triplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO: Henkel Corporation, Law Dept.
2500 Renaissance Blvd, Suite 200
Gulph Mills, PA 19406

SIGNATURE

Glenn E. J. Murphy
NAME ATTORNEY FOR APPLICANT

33,539
REGISTRATION NUMBER

Form PTO 1390 (REV 5-93)

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PATENT
Docket H 3763 PCT/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Re: PCT/EP99/09683

International Filing Date:	December 9, 1999
Priority Date:	December 18, 1998
Applicant:	KROPF, et al.
Title:	FINE SUSPENSIONS OF POORLY SOLUBLE CALCIUM SALTS AND THEIR USE IN DENTAL CARE PRODUCTS

PRELIMINARY AMENDMENT

Assistant Commissioner of Patents
Washington, DC 20231

Please enter the amendments below before examining this case on the merits:

IN THE SPECIFICATION:

On page 1, insert below the title:

--CROSS-REFERENCE TO RELATED APPLICATIONS

This application is the U.S. National Stage application filed under 35 U.S.C. § 371 of International Application No. PCT/EP99/09683, filed December 9, 1999, in the European Patent Office, claiming priority under 35 U.S.C. §§ 119 and 365 of PCT/EP99/09683 and DE 198 58 662.0, filed on December 18, 1998, in the German Patent Office.--

On page 3, after line 30, insert the heading --DESCRIPTION OF THE INVENTION--.

IN THE CLAIMS:

Please cancel claims 1 to 7 without prejudice, and add new claims 8 to 14:

8. A suspension of one or more phosphate, fluoride, or fluorophosphate calcium salts in a liquid medium in which the salts are less than 1 g/l soluble, wherein the calcium salts comprise primary particles having diameters of from 5 to 50 nanometers and lengths of from 10 to 150 nanometers, stabilized against agglomeration by a content of at least 0.01% by weight, based on the weight of the suspension, of a water-soluble surfactant or of a water-soluble polymeric protective colloid.

9. The suspension of claim 8, comprising 1% to 40% by weight of the one or more calcium salts and 0.1% to 10% by weight, based on the weight of the one or more calcium salts, of the water-soluble surfactant or the water-soluble polymeric protective colloid.

10. The suspension of claim 9, comprising 1% to 10% by weight, based on the weight of the one or more calcium salts, of one or more nonionic surfactants.

11. A process for the preparation a suspension of poorly soluble calcium salts, comprising the steps of precipitating one or more phosphate, fluoride, or fluorophosphate calcium salts in an aqueous medium in which these salts are less than 1 g/l soluble, wherein the calcium salts comprise primary particles having diameters of from 5 to 50 nanometers and lengths of from 10 to 150 nanometers, said precipitation being carried out in the presence of water-

soluble surfactants or water-soluble polymeric protective colloids.

12. The process of claim 11, wherein the aqueous medium is an acidic solution of a water-soluble calcium salt and a stoichiometric amount of a water-soluble phosphate salt with a pH below 3, and the precipitation is effected by increasing the pH using aqueous alkalis or ammonia in the presence of the water-soluble surfactants or water-soluble polymeric protective colloids.

13. A toothpaste comprising one or more silica polishing agents, humectants, binders or aromas and 0.1-5% by weight of one or more calcium salts selected from the group consisting of amorphous calcium phosphate, hydroxylapatite, fluorapatite, and calcium fluoride, said calcium salts being present in the form of a suspension of one or more of the salts in a liquid medium in which the salts are less than 1 g/l soluble, wherein the salts comprise primary particles having diameters of from 5 to 50 nanometers and lengths of from 10 to 150 nanometers, stabilized against agglomeration by a content of at least 0.01% by weight, based on the weight of the suspension, of a water-soluble surfactant or of a water-soluble polymeric protective colloid.

14. A method of remineralizing teeth comprising the steps of applying to a tooth a remineralizing-effective amount of a suspension of one or more phosphate, fluoride, or fluorophosphate calcium salts in a liquid medium in which the salts are less than 1 g/l soluble, wherein the calcium salts comprise primary particles having diameters of from 5 to 50 nanometers and lengths of from 10 to 150 nanometers, stabilized against agglomeration by a content of at least

0.01% by weight, based on the weight of the suspension, of a water-soluble surfactant or of a water-soluble polymeric protective colloid.

REMARKS

Claims 1-7 have been canceled without prejudice, and new claims 8-14 added. The subject matter of the new claims is described in the specification at page 3, line 32 to page 4, line 12, page 7, lne 19, to page 8, line 4, page 8, lines 17-26, and page 10, lines 4-9, as well as in the claims as originally filed. The specification has been amended to include a cross-reference to related applications and headings appropriate to U.S. practice. No new matter has been added.

The new claims better claim the full literal and equivalent scope and breadth of subject matter disclosed in the application, notwithstanding applicants' belief that the original claims, drafted for examination in the German and European Patent Offices, would have been allowable but for minor matters of form permitted in German or European practice but objected to in the U.S.P.T.O. The new claims find support in the application independent of the original claims and therefore are not believed to constitute narrowing amendments to the original claims within the holding of Festo Corp. v. Shoketsu Kinzoku Kogyo Kabushiki Co., No. 95-1066 (Fed. Cir. Nov. 29, 2000).

Applicants respectfully request entry of this Amendment and examination of the application. If any fees are due to enter this paper that have not been accounted for, please charge Deposit Account No. 01-1250.

Respectfully submitted,

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Patent Application

"Fine suspensions of poorly soluble calcium salts and their use in dental care products"

The invention relates to fine suspensions of poorly soluble calcium salts which, because of their particle size in the nanometer range and their stability toward agglomeration, are particularly suitable for use in dental care products.

Phosphate salts of calcium have for a long time been added either as abrasive components or to promote remineralization of tooth enamel to formulations of dental cleaning products and dental care products. This is true particularly for hydroxylapatite and fluorapatite, and for amorphous calcium phosphates and for brushite (dicalcium phosphate dihydrate). However, calcium fluoride has also been described a number of times as a constituent of dental cleaning products and as a component for strengthening tooth enamel and for the prophylaxis of caries.

The availability of these substances for the desired remineralization depends quite decisively on the particle size of these poorly water-soluble components dispersed in the dental care products. It has therefore been proposed to use these poorly soluble calcium salts in extremely fine dispersion.

DE-A-2134862 discloses, for example, a dental care product for hypersensitive teeth which comprises very finely divided hydroxylapatite ($Ca_5[(PO_4)_3OH]$) whose particle size, however, is given as 6-8 µm (micrometers) since greater finenesses cannot be achieved by grinding.

Dental care products comprising separate components have also already been proposed, of which one comprises a dissolved Ca salt and the other comprises a dissolved phosphate or fluoride salt, and which are combined only shortly prior to application - or which are used in succession - in order to apply the freshly precipitated and still amorphous or finely crystalline calcium salts to the tooth surface. The disadvantages of such handling are obvious since the user has to use two products successively or combine them shortly before use. If compositions which comprise freshly precipitated, still amorphous calcium phosphates or calcium fluoride are stored, the precipitates age, the crystallites grow and agglomerate to give coarser secondary particles. This reduces the remineralizing action and jeopardizes the stability of the dispersion.

The object was therefore to provide suspensions of such poorly soluble calcium salts whose particle size is in the nanometer range and which are largely protected against agglomeration.

WO 94/04460 A1 describes a process for the preparation of amorphous calcium salts and their use for the remineralization of teeth. EP 786245 A1 describes dental care products which comprise hydroxylapatite having particle sizes of from 0.05 to 1.0 µm which are obtained by grinding. WO 98/18719 discloses a hydroxylapatite composition which comprises hydroxylapatite with particle diameters of 10-20 nm and particle lengths of 50-100 nm and which are intended to be used, for example, in toothpastes. These are obtained by concentrating very dilute suspensions by two or more filtration steps.

EP 0499299 A2 discloses suspensions of particles of crystalline drugs which have a size of less than 100 nm and contain, adsorbed on their surface, a surface modifier which may also be a surfactant or a polymeric protective colloid. Stabilization of inorganic poorly soluble salts obtained by precipitation reactions is not disclosed. WO 96/34829 A1 discloses a process for the preparation of little-agglomerated particles in the nanometer range, in which a suspension of such particles is prepared from the precursors in a liquid medium which has no noteworthy solvency for the particles, in the presence of a surface-blocking substance. In another embodiment, a sol which comprises amorphous or partially crystalline nanoparticles is suspended in the presence of the surface-blocking substance. Also named as surface-blocking substances are (poly)carboxylic acids and nonionogenic surfactants. Disclosed as suitable particles are, however, only oxide (hydrates), sulfides, selenides, tellurides and phosphides precipitated from hydrolyzable salts or organometallic compounds by adding water or changing the pH. Phosphates or fluorides of calcium or use of the suspensions in dental care products are not disclosed.

It has now been found that suspensions of poorly water-soluble calcium salts in very finely divided form can be stabilized during the precipitation or shortly thereafter if the precipitation is carried out in the presence of an agglomeration inhibitor, or the dispersion is redispersed in the presence of the agglomeration inhibitor.

The invention therefore provides a suspension of poorly water-soluble calcium salts, chosen from phosphates, fluorides and fluorophosphates, in a liquid medium in which these calcium salts are insoluble or poorly

soluble, characterized in that the calcium salts are present in the form of primary particles having diameters of from 5 to 50 nanometers and lengths of from 10 to 150 nanometers and are stabilized against agglomerization by a content of at least 0.01% by weight, based on the weight of the suspension, of a water-soluble surfactant or of a water-soluble polymeric protective colloid.

Poorly soluble or poorly water-soluble salts are to be understood as meaning those salts which are soluble in water or in the liquid suspension medium to an amount of less than 1 g/l (20°C). Suitable salts are preferably calcium hydroxyphosphate ($Ca_5[OH(PO_4)_3]$) or hydroxylapatite, calcium fluorophosphate, ($Ca_5[F(PO_4)_3]$) or fluorapatite, F-doped hydroxylapatite of the general composition $Ca_3(PO_4)_3(OH,F)$ and calcium fluoride (CaF_2) or fluorite (fluorspar).

A suitable liquid medium in which the calcium salts can be dispersed is primarily water. However, the calcium salt particles isolated from an aqueous suspension, e.g. by filtration or centrifugation, can also be redispersed in organic solvents and, in this case, likewise produce suspensions of the primary particles in the nanometer range which have virtually no tendency for agglomeration. Suitable organic liquid media are, for example, water-soluble, lower alcohols and glycols, polyethylene glycols, glycerol or mixtures thereof with one another or with water.

Primary particles are understood here as meaning the crystallites, i.e. the individual crystals, of said calcium salts. The particle diameter should be understood here as meaning the smallest diameter, and the length to be understood as meaning the greatest diameter of the

crystal particles, e.g. the length of a rod-shaped crystallite. Wherever an average particle diameter is discussed, this is understood as meaning a volume-averaged value.

For the purposes of the present invention, water-soluble surfactants are understood as meaning all surface-active substances characterized by a lipophilic alkyl, alkylphenyl or acyl radical having 8-22 carbon atoms and a hydrophilic, ionic or nonionic group which imparts to the surfactant a solubility in water of more than 1 g/l (20°C). Suitable as anionic surfactants are, for example, the alkali metal or ammonium salts of C_8-C_{18}-alkanecarboxylic acids (soaps), of alkyl-(C_{12}-C_{18}) sulfuric monoesters (alkyl sulfates), of alkylpolyglycol ether sulfuric monoesters (ether sulfates), of sulfosuccinic mono-C_8-C_{18}-alkyl esters (sulfosuccinates), of alkanesulfonic acids (alkanesulfonates), of C_{12}-C_{18}-acyloxyethanesulfonic acids (isethionates), of C_{12}-C_{18}-acylaminoalkanesulfonic acids (taurides), of N-C_{12}-C_{18}-acylsarcosine (sarcosinates), of alkylpolyglycol ether carboxylic acids (ether carboxylates), of alkyl(polyglycol ether) phosphoric acids (alkyl(polyglycol ether) phosphate).

Suitable cationic surfactants are, for example, alkyltrimethylammonium chloride, alkyldimethylbenzyl-ammonium chloride, alkylpyridinium chloride, alkyldimethylhydroxyethylammonium chloride, acylimidazolinium methosulfates and acyloxyethyltrimethylammonium chloride.

Suitable zwitterionic surfactants are, for example, betaine surfactants, such as, for example,

alkyldimethylcarboxymethylbetaine and acylaminoalkyldimethylcarboxymethylbetaine.

Amphoteric surfactants, such as, for example, alkylaminopropanecarboxylic acids, are also suitable as ionic surfactants.

However, the nonionic surfactants are preferably suitable, in particular the addition products of ethylene oxide to lipids with mobile hydrogen atoms. Such suitable nonionic surfactants are, for example, the addition products of 6-60 mol of ethylene oxide to linear fatty alcohols, to fatty acids, to fatty amines, to fatty acid monoglycerides, to sorbitan fatty acid monoesters, to alkylphenols, to sugar fatty acid monoesters, to methylglucoside fatty acid monoesters and to fatty acid monoethanolamides. Further preferably suitable nonionic surfactants are the alkyl (oligo) glucosides obtainable by reacting glucose with C_8-C_{18}-fatty alcohols or by transacetylation of butyl(oligo) glucoside with fatty alcohols. Preferably suitable alkyl (oligo) glucosides are, for example, the alkyl (C_8-C_{16}) glucosides having average degrees of oligomerization (of the glucoside radical) of from 1 to 2. Such products are [lacuna] commercially, e.g. under the trade name Plantacare® 1200 or Plantacare® 600. Further preferably suitable nonionogenic surfactants are the mixtures obtainable by ethoxylation of hydrogenated castor oil which are obtained, for example, by the addition of 30, 40 or 60 mol of ethylene oxide to hydrogenated castor oil.

Finally, amine oxide surfactants and sugar fatty acid esters are also suitable as nonionogenic surfactants.

Water-soluble polymeric protective colloids are understood as meaning high molecular weight compounds which are adsorbed on the surface of the nanoparticles and modify these such that they are hindered from coagulating and agglomerating. Suitable polymeric protective colloids are, for example, natural water-soluble polymers, such as, for example, gelatin, casein, albumin, starch, plant gums and water-soluble derivatives of water-insoluble polymeric natural substances, such as, for example, cellulose ethers (methylcellulose, hydroxyethylcellulose, carboxymethylcellulose), hydroxyethylstarch or hydroxypropylguar.

Synthetic water-soluble polymers suitable as protective colloids are, for example, polyvinyl alcohol, polyvinylpyrrolidone, polyacrylic acids, polyaspartic acid and others.

The suspensions according to the invention are prepared by precipitation reactions from aqueous solutions of water-soluble calcium salts and aqueous solutions of water-soluble phosphate or fluoride salts. Here the precipitation is carried out in the presence of water-soluble surfactants or water-soluble polymeric protective colloids. This may, for example, be carried out by adding the surfactants or protective colloids to the aqueous phosphate or fluoride salt solution or to the solution of the calcium salt prior to the reaction. Alternatively, the aqueous calcium salt solution can be added to an aqueous surfactant or protective colloid solution at the same time as the phosphate or fluoride salt solution.

A further process variant involves the precipitation being carried out from a strongly acidic solution of a water-soluble calcium salt and a stoichiometric amount of

a water-soluble phosphate salt with a pH below 3 by increasing the pH using an aqueous alkali or ammonia in the presence of water-soluble surfactants or water-soluble polymeric protective colloids.

The concentration of poorly soluble calcium salt in the suspensions according to the invention can cover a wide range from about 1 to 40% by weight. Here, the content can be increased on the one hand during the preparation by means of the concentration of the water-soluble salts, and on the other hand after the precipitation reaction by concentration, e.g. by filtration or centrifugation or by distilling off some of the water, without the effect of the surfactant or of the protective colloid being lost in the process.

The concentration of the surfactant or of the polymeric protective colloid in the aqueous suspension is, for example, 0.1 to 20% by weight, preferably 0.1-10% by weight, based on the content of poorly soluble calcium salt. In a preferred embodiment, the suspension according to the invention therefore comprises 1-40% by weight of the poorly soluble calcium salts and, for the stabilization, 0.1-10% by weight of a water-soluble surfactant or of a water-soluble polymeric protective colloid, based on the weight of the calcium salt.

Preferably suitable for the stabilization against agglomeration are predominantly the nonionic surfactants in an amount of from 1 to 10% by weight, based on the weight of the calcium salt. The nonionic surfactants of the type of alkyl C_8-C_{16}-(oligo)- glucosides and of ethoxylates of hydrogenated castor oil have proven particularly effective. These can also be used together

with the polymeric protective colloids for the stabilization.

For the preparation of suspensions according to the invention in other liquid media, it is expedient to start from aqueous suspensions according to the invention, free these by filtration or centrifugation from the aqueous phase, dry, where appropriate, the nanoparticles and redisperse them in organic solvents. Here, a fresh addition of surfactants or protective colloids is no longer necessary since the nanoparticles comprise the amounts of stabilizer required for inhibition of agglomeration adsorbed on the surface. The finely divided nature and stability of such suspensions is therefore comparable with those of the aqueous suspensions. Another possibility consists in mixing the aqueous suspension with a higher-boiling solvent, e.g. with glycerol, and removing the water by distillation. Suitable as organic liquid medium is, particularly with regard to use in dental care products, primarily glycerol and its liquid mixtures with sorbitol and optionally with water.

The suspensions according to the invention, in particular those of hydroxylapatite, fluorapatite and calcium fluoride, are suitable as remineralizing component for the preparation of compositions for the cleaning and care of teeth. As a result of the particularly finely divided nature, the effect, known per se, of strengthening the tooth enamel and closing lesions and dentinal tubules can take place particularly rapidly and completely. The compositions for the cleaning and care of teeth may here be in the form of pastes, liquid creams, gels or mouthwashes. Even in liquid preparations, the suspensions according to the invention disperse readily and the

calcium salts remain stably dispersed and do not tend toward sedimentation.

A preferred embodiment are, however, toothpastes with a content of silica, polishing agents, humectants, binders and aromas which comprise 0.1-5% by weight of finely divided calcium salts from the group hydroxylapatite, fluorapatite and calcium fluoride in the form of a suspension according to the invention.

The preparations for the cleaning and care of teeth can comprise the customary components and auxiliaries of such compositions in the amounts customary for this purpose. For toothpastes, these are, for example,

- cleaning and polishing substances, such as, for example, chalk, silicas, aluminum hydroxide, aluminum silicates, calcium pyrophosphate, dicalcium phosphate, insoluble sodium metaphosphate or synthetic-resin powder
- humectants, such as, for example, glycerol, 1,2-propylene glycol, sorbitol, xylitol and polyethylene glycols
- binders and consistency regulators, e.g. natural and synthetic water-soluble polymers and water-soluble derivatives of natural substances, e.g. cellulose ethers, phyllosilicates, finely divided silicas (aerogel silicas, pyrogenic silicas)
- aromas, e.g. peppermint oil, spearmint oil, eucalyptus oil, aniseed oil, fennel oil, caraway oil, menthyl acetate, cinnamaldehyde, anethole, vanillin, thymol and mixtures of these and other natural and synthetic aromas
- sweeteners, such as, for example, saccharin-sodium, sodium cyclamate, aspartame, acesulfame K, stevioside,

monellin, glycyrrhicine, dulcin, lactose, maltose or fructose

- preservatives and antimicrobial substances, such as, for example, p-hydroxybenzoates, sodium sorbate, triclosan, hexachlorophene, phenylsalicylates, thymol etc.
- pigments, such as, for example, titanium dioxide or pigment dyes for producing colored stripes
- buffer substances, e.g. primary, secondary or tertiary alkali metal phosphates, citric acid/Na citrate
- wound-healing and antiinflammatory active ingredients, e.g. allantoin, urea, azulene, panthenol, acetylsalicylic acid derivatives, plant extracts, vitamins, e.g. retinol or tocopherol.

The examples below serve to illustrate the subject-matter of the invention in more detail:

Examples

1. Preparation of suspensions of poorly soluble calcium salts

1.1 Preparation of a hydroxylapatite suspension by precipitation and redispersion

50.86 g of $Ca(NO_3)_2 \cdot 4H_2O$ were dissolved in demin. water and made up to 200 ml. 10 g of Plantacare 1200® were added to this. 60 ml of 25% strength ammonia solution were then added, so that the pH was 12.

17 g of ammonium hydrogenphosphate with dissolved in demin. water and made up to 200 ml. 10 g of Plantacare 1200® were added to this. 60 ml of 25% strength ammonia solution were then added.

Both solutions were brought to 75°C and mixed with vigorous stirring. After stirring for one hour, the precipitate was centrifuged off, washed a number of times with water and then taken up in water to give a 5% strength by weight hydroxylapatite suspension. The particle sizes were 4-10 nm x 60-130 nm (diameter x length).

(demin. = demineralized)

1.2 Preparation of a hydroxylapatite suspension by reprecipitation (pH shift) and concentration by evaporation

25.43 g of $Ca(NO_3)_2 \cdot 4H_2O$ were dissolved in demin. water and made up to 100 ml. 8.5 g of ammonium hydrogenphosphate were likewise dissolved in demin. water and made up to 100 ml. The solutions were combined, with formation of a voluminous precipitate. 37% strength hydrochloric acid was added dropwise to the suspension until the precipitate had completely dissolved at pH 2.

A mixture of 200 ml of demin. water, 200 ml of 25% strength ammonia solution and 20 g of Cremophor RH 60® (BASF, castor oil + 60 EO) was initially introduced. At 0°C, the apatite solution was added dropwise to this solution with stirring, with formation of a precipitate. Excess ammonia was separated off by distillation, then the mixture was washed by means of dialysis until nitrate-free. Concentration by evaporation on a rotary evaporator gave a 10% strength by weight suspension of hydroxylapatite. The particle sizes were 30 nm (volume-averaged) in diameter (determination using a Micro-Trac 3.150 Ultrafine Particle Analyzer 150 by averaging over the total particle volume).

1.3 Preparation of a suspension of hydroxylapatite analogously to Example 1.2 (starting from $CaCl_2$)

11.95 g of calcium chloride were dissolved in demin. water and made up to 100 ml. 7.4 g of ammonium hydrogenphosphate were likewise dissolved in demin. water and made up to 100 ml. The solutions were combined with formation of a voluminous precipitate. 37% strength hydrochloric acid was added dropwise to the suspension until the precipitate had completely dissolved at pH 2.

A mixture of 200 ml of demin. water, 200 ml of 25% strength ammonia solution and 20 g of Cremophor RH 60® (BASF, castor oil + 60 EO) was initially introduced. At 0°C, the apatite solution was added dropwise to this solution with stirring, with formation of a precipitate. Excess ammonia was separated off by distillation, then the mixture was washed by means of dialysis until nitrate-free. Concentration by evaporation on a rotary evaporator gave a 10% strength by weight suspension of hydroxylapatite. The particle sizes were 10-35 nm × 20-50 nm (diameter × length).

1.4 Preparation of the hydroxylapatite suspension analogously to Example 1.2 using Arlatone 289 (BASF)

Instead of 20 g of Cremophor RH 60, 35 g of Arlatone 289 were used. A 10% strength by weight suspension of hydroxylapatite with an average particle size of 40 nm was obtained. (Micro-Trac 3.150 Ultrafine Particle Analyzer).

1.5 Preparation of a hydroxylapatite suspension in glycerol

0.3 mol of calcium chloride were dissolved in 2000 ml of demin. water and thermostatted at 25°C. Ammonia solution was used to establish a pH of 12. Then, with vigorous stirring, a solution of 0.18 mol of ammonium hydrogenphosphate in 400 ml of demin. water, which was thermostatted at 25°C and had been adjusted to pH 10 using ammonia, was slowly added dropwise. After a reaction time of 20 h, 3 g of Cremophor RH 60® solution (40% strength by weight in demin. water) were added and dispersed by inputting chemical energy (stirring, ultrasound). The suspension was then centrifuged off a number of times and washed firstly with 1% strength aqueous Cremophor RH60® solution, then with ethanol. The material was then taken up in 100 ml of glycerol. Hydroxylapatite particles with sizes of 5-20 nm x 10-70 nm (diameter x length) were present in this glycerol suspension.

1.6 Preparation of a suspension of fluorine-doped hydroxylapatite in glycerol

0.3 mol of calcium chloride were dissolved in 2000 ml of demin. water and thermostatted at 25°C. Ammonia solution was used to establish a pH of 12. For this, a solution of 2.27 g of ammonium fluoride in 50 ml of demin. water was added. Then, with vigorous stirring, a solution of

0.18 mol of ammonium hydrogenphosphate in 400 ml of demin. water, which was thermostatted at 25°C and had been adjusted to pH 10 using ammonia, was slowly added dropwise. After a reaction time of 20 h, 3 g of Cremophor RH 60® solution (40% strength by weight in demin. water) were added and dispersed by inputting chemical energy (stirring, ultrasound). The suspension was then centrifuged off a number of times and washed firstly with 1% strength aqueous Cremophor RH60® solution, then with ethanol. The material was then taken up in 100 ml of glycerol. Here, a glycerol suspension of $Ca_5(PO_4)_3(OH,F)$ particles with a size of 5-20 nm × 10-70 nm (diameter × length) was obtained.

1.7 Preparation of a calcium fluoride suspension by precipitation

11.95 g of anhydrous $CaCl_2$ were dissolved in demin. water made up to 100 ml. 200 ml of demin. water, 35 g of Arlatone 289 (BASF) and 15 g of ammonium fluoride were mixed in a receiver. Both solutions were cooled to 0°C and the first solution was added to the second with vigorous stirring. The dispersion formed was concentrated by evaporation on a rotary evaporator at 70°C until the solids content was 10% by weight. Washing was then carried out by means of dialysis. This gave a calcium fluoride suspension with an average (volume-weighted) particle size of 20 nm.

2. Dental creams with calcium salt nanoparticles

Formulation examples	2.1	2.2
Sident® 8	10.0% by wt.	10.0% by wt.
Sident® 22S	7.0% by wt.	7.0% by wt.
Sipernat® 320DS	0.8% by wt.	0.8% by wt.
CaF_2 suspension Example 1.7	5.0% by wt.	-
Hydroxylapatite suspension Example 1.1	-	5.0% by wt.
Polywax 1550	2.0% by wt.	2.0% by wt.
Texapon K 1296	1.5% by wt.	1.5% by wt.
Titanium dioxide	1.0% by wt.	1.0% by wt.
Cekol 500 T	1.0% by wt.	1.0% by wt.
Na fluoride	0.33% by wt.	0.33% by wt.
Na benzoate	0.25% by wt.	0.25% by wt.
Aroma	1.0% by wt.	1.0% by wt.
Tagat S	0.2% by wt.	-
Na saccharinate	0.15% by wt.	0.15% by wt.
Trisodium phosphate	0.10% by wt.	0.10% by wt.
Sorbitol (70% strength in water)	31.0% by wt.	31.0% by wt.
Water	ad 100% by wt.	ad 100% by wt.

The following commercial products were used:

Plantaren® 1200: C_{12}-C_{16}-fatty alcohol oligo-(1.4)-glucoside about 50% by weight in water
Manufacturer: HENKEL KGaA

Cremophor® RH 60: Castor oil (hydrogenated) poly(60)-glycol ether
Manufacturer: BASF

Arlatone® 289: Castor oil (hydrogenated) poly(54)-glycol ether
Manufacturer: Atlas Chemie (ICI)

Sident® 8: Synth. amorph. silica, BET 60 m^2/g
Tamped density: 350 g/l
Manufacturer: DEGUSSA

Sident® 22 S: Hydrogel silica, BET 140 m^2/g
Tamped density: 100 g/l
Manufacturer: DEGUSSA

Polywax® 1550: Polyethylene glycol, MW: 1550
Softening point 45-50°C
Manufacturer: RWE/DEA

Texapon® K 1296: Sodium lauryl sulfate powder
Manufacturer: HENKEL KGaA

Cekol® 500 T: Sodium carboxymethylcellulose
Viscosity (2% strength in water, Brookfield LVF 20°C): 350-700 mPas
Supplier: Nordmann-Rassmann

Tagat® S: Polyoxyethylene-(20) glyceryl monostearate
Manufacturer: Tego Cosmetics (Goldschmidt)

Patent claims

1. A suspension of poorly water-soluble calcium salts, chosen from phosphates, fluorides and fluorophosphates, in a liquid medium in which these salts are insoluble or poorly soluble, characterized in that the calcium salts are present in the form of primary particles having diameters of from 5 to 50 nanometers and lengths of from 10 to 150 nanometers and are stabilized against agglomerization by a content of at least 0.01% by weight, based on the weight of the suspension, of a water-soluble surfactant or of a water-soluble polymeric protective colloid.

2. The suspension as claimed in claim 1, characterized in that 1 to 40% by weight of the poorly soluble calcium salts and, for the stabilization, 0.1 to 10% by weight, based on the weight of the poorly soluble calcium salt, of a water-soluble surfactant or of a water-soluble polymeric protective colloid are present in the suspension.

3. The suspension as claimed in claim 1 or 2, characterized in that, for the stabilization, nonionic surfactants are present in an amount of from 1 to 10% by weight, based on the weight of the poorly soluble calcium salt.

4. A process for the preparation of the suspension as claimed in claim 1-3 by precipitation processes from aqueous solutions of water-soluble calcium salts and aqueous solutions of water-soluble phosphate or fluoride salts, characterized in that the precipitation is carried out in the presence of

water-soluble surfactants or water-soluble polymeric protective colloids.

5. A process for the preparation of the suspension as claimed in claim 1-3 by precipitation from an acidic solution of a water-soluble calcium salt and a stoichiometric amount of a water-soluble phosphate salt with a pH below 3 by increasing the pH using aqueous alkalis or ammonia in the presence of water-soluble surfactants or water-soluble polymeric protective colloids.

6. The use of the suspension as claimed in any of claims 1-3 as remineralizing component in compositions for the cleaning and care of teeth.

7. A toothpaste with a content of silica polishing agents, humectants, binders and aromas, characterized in that 0.1-5% by weight of fine calcium salts from the group amorphous calcium phosphate, hydroxylapatite, fluorapatite and calcium fluoride are present in the form of a suspension as claimed in claim 1-3.

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AUG 15 2001

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Rev. 8/95

U.S. Department of Commerce
Patent and Trademark Office

DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION

Attorney Docket Number	H 3763 PCT/US
First Named Inventor	KROPF, Christian
COMPLETE IF KNOWN	
Application Number	
Filing Date	
Group Art Unit	
Examiner Name	

☐ Declaration Submitted with Initial Filing OR ☒ Declaration Submitted after Initial Filing

As a below named inventor, I hereby declare that:
My residence, post office address, and citizenship are as stated below next to my name.
I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

FINE SUSPENSIONS OF POORLY SOLUBLE CALCIUM SALTS AND THEIR USE IN DENTAL CARE PRODUCTS

(Title of the Invention)

the specification of which

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY) 12/19/1999 as United States Application Number or PCT International Application Number PCT/EP99/09683 and was amended on (MM/DD/YYYY) _____ (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY	Priority Not Claimed	Certified Copy Attached? YES	Certified Copy Attached? NO
198 58 662.0	Germany	12/18/1998	☐	☐	☒

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I hereby claim the benefit under Title 35, United States Code §119(e) of any United States provisional application(s) listed below.

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U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number *(if applicable)*
	PCT/EP99/09683	12/09/1999	

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As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

☐ Firm Name _____ Customer Number or label _____

OR

☒ List Attorney(s) and/or agent(s) name and registration number below:

Name	Registration Number	Name	Registration Number
Wayne C. Jaeschke	21,062	Glenn E. J. Murphy	33,539
Kimberly R. Hild	39,224	Stephen D. Harper	33,243

☐ Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

Please direct all correspondence to: ☒ Customer Number or label 00423 OR ☐ Fill in correspondence address below

Name	Glenn E. J. Murphy				
Address	Henkel Corporation - Patent Department				
Address	2500 Renaissance Boulevard, Suite 200				
City	Gulph Mills	State	PA	ZIP	19406
Country	USA	Telephone	610-278-4926	Fax	610-278-6548

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor: ☐ A petition has been filed for this

Given Name	**Christian**	Middle Initial		Family Name	**KROPF**	Suffix e.g. Jr.	
Inventor's Signature				Date			
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Post Office Address							
City	**40597 Duesseldorf**	State		Zip		Country	**Germany**
Applicant Authority							

☒ Additional inventors are being named on supplemental sheet(s) attached hereto

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H 3763 PCT/US

DECLARATION

ADDITIONAL INVENTOR(S)
Supplemental Sheet

Name of Additional Joint Inventor, if any: [ ] A petition has been filed for this unsigned inventor

Given Name	Ulrike	Middle Initial		Family Name	BRUENINGHAUS	Suffix e.g. Jr.	
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Post Office Address							
City	40789 Monheim	State		Zip		Country	Germany	Applicant Authority	

Name of Additional Joint Inventor, if any: [ ] A petition has been filed for this unsigned inventor

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Inventor's Signature						Date	
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Post Office Address							
City	58453 Witten	State		Zip		Country	Germany	Applicant Authority	

Name of Additional Joint Inventor, if any: [ ] A petition has been filed for this unsigned inventor

Given Name	Michael	Middle Initial		Family Name	MEINDERS	Suffix e.g. Jr.	
Inventor's Signature						Date	
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Post Office Address							
City	47800 Krefeld	State		Zip		Country	Germany	Applicant Authority	

Name of Additional Joint Inventor, if any: [ ] A petition has been filed for this unsigned inventor

Given Name	Peter	Middle Initial		Family Name	WUELKNITZ	Suffix e.g. Jr.	
Inventor's Signature						Date	
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Post Office Address	Im Erlengrund 9						
Post Office Address							
City	42799 Leichlingen	State		Zip		Country	Germany	Applicant Authority	

[X] Additional inventors are being named on supplemental sheet(s) attached hereto

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Patent and Trademark Office

DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION

☐ Declaration Submitted with Initial Filing OR ☒ Declaration Submitted after Initial Filing

Attorney Docket Number	H 3763 PCT/US
First Named Inventor	KROPF, Christian
COMPLETE IF KNOWN	
Application Number	
Filing Date	
Group Art Unit	
Examiner Name	

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I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

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(Title of the Invention)

the specification of which

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY) 12/19/1999 as United States Application Number or PCT International Application Number PCT/EP99/09683 and was amended on (MM/DD/YYYY) _____ (if applicable).

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☐ Firm Name _____ Customer Number or label _____

OR

☒ List Attorney(s) and/or agent(s) name and registration number below:

Name	Registration Number	Name	Registration Number
Wayne C. Jaeschke	21,062	Glenn E. J. Murphy	33,539
Kimberly R. Hild	39,224	Stephen D. Harper	33,243

☐ Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

Please direct all correspondence to: ☒ Customer Number or label 00423 OR ☐ Fill in correspondence address below

Name	Glenn E. J. Murphy						
Address	Henkel Corporation - Patent Department						
Address	2500 Renaissance Boulevard, Suite 200						
City	Gulph Mills			State	PA	ZIP	19406
Country	USA	Telephone	610-278-4926		Fax	610-278-6548	

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor: ☐ A petition has been filed for this

Given Name	**Christian**	Middle Initial		Family Name	**KROPF**	Suffix e.g. Jr.	
Inventor's Signature					Date		
Residence: City	**Duesseldorf**	State		Country	**Germany**	Citizenship	**Germany**
Post Office Address	**Caecilienstrasse 4**						
Post Office Address							
City	**40597 Duesseldorf**	State		Zip		Country	**Germany**	Applicant Authority	

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DECLARATION

ADDITIONAL INVENTOR(S) Supplemental Sheet

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Ulrike	Middle Initial		Family Name	BRUENINGHAUS	Suffix e.g. Jr.	
Inventor's Signature						Date	
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Post Office Address							
City	40789 Monheim	State		Zip		Country	Germany
Applicant Authority							

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Given Name	Amergio	Middle Initial		Family Name	PASTURA	Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City	Witten	State		Country	Germany	Citizenship	Germany
Post Office Address	Sauerbruchstr. 3a						
Post Office Address							
City	58453 Witten	State		Zip		Country	Germany
Applicant Authority							

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Michael	Middle Initial		Family Name	MEINDERS	Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City	Krefeld	State		Country	Germany	Citizenship	Germany
Post Office Address	Am Eickerhof 11						
Post Office Address							
City	47800 Krefeld	State		Zip		Country	Germany
Applicant Authority							

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Peter	Middle Initial		Family Name	WUELKNITZ	Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City	Leichlingen	State		Country	Germany	Citizenship	Germany
Post Office Address	Im Erlengrund 9						
Post Office Address							
City	42799 Leichlingen	State		Zip		Country	Germany
Applicant Authority							

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DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION

☐ Declaration Submitted with Initial Filing OR ☒ Declaration Submitted after Initial Filing

Attorney Docket Number	H 3763 PCT/US
First Named Inventor	KROPF, Christian
COMPLETE IF KNOWN	
Application Number	
Filing Date	
Group Art Unit	
Examiner Name	

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I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

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(Title of the Invention)

the specification of which

☐ is attached hereto

OR

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ADDITIONAL INVENTOR(S) Supplemental Sheet

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Rolf	Middle Initial		Family Name	HEMPELMANN	Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City	St. Ingbert	State		Country	Germany	Citizenship	Germany
Post Office Address	St. Herblainer Str. 11						
Post Office Address							
City	66386 St. Ingbert	State		Zip		Country	Germany	Applicant Authority	

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Marcel	Middle Initial		Family Name	ROTH	Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City	Duesseldorf	State		Country	Germany	Citizenship	Germany
Post Office Address	Weststrasse 17						
Post Office Address							
City	40597 Duesseldorf	State		Zip		Country	Germany	Applicant Authority	

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name		Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		Zip		Country		Applicant Authority	

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name		Suffix e.g. Jr.	
Inventor's Signature						Date	
Residence: City		State		Country		Citizenship	
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☐ Firm Name | Customer Number or label

OR

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Name	Registration Number	Name	Registration Number
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Kimberly R. Hild	39,224	Stephen D. Harper	33,243

☐ Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

Please direct all correspondence to: ☒ Customer Number or label 00423 OR ☐ Fill in correspondence address below

Name	Glenn E. J. Murphy				
Address	Henkel Corporation - Patent Department				
Address	2500 Renaissance Boulevard, Suite 200				
City	Gulph Mills	State	PA	ZIP	19406
Country	USA	Telephone	610-278-4926	Fax	610-278-6548

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor: ☐ A petition has been filed for this

Given Name	Christian	Middle Initial		Family Name	KROPF	Suffix e.g. Jr.	
Inventor's Signature	Christian Kropf			Date	August 1, 2001		
Residence: City	Duesseldorf DEX	State		Country	Germany	Citizenship	Germany
Post Office Address	Caecilienstrasse 4						
Post Office Address							
City	40597 Duesseldorf	State		Zip		Country	Germany
Applicant Authority							

☒ Additional inventors are being named on supplemental sheet(s) attached hereto

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DECLARATION

ADDITIONAL INVENTOR(S) Supplemental Sheet

2-00

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Ulrike	Middle Initial		Family Name	BRUENINGHAUS	Suffix e.g. Jr.	
Inventor's Signature	Ulrike Brueninghaus			Date	August 1, 2001		
Residence: City	Monheim OEX	State		Country	Germany	Citizenship	Germany ✓
Post Office Address	An der Dorfstr. 6						
Post Office Address							
City	40789 Monheim	State		Zip		Country	Germany
Applicant Authority							

3-00

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	~~Amergio~~ Amerigo 25.7.01 Pa	Middle Initial		Family Name	PASTURA	Suffix e.g. Jr.	
Inventor's Signature	Amerigo Pastura			Date	August 1, 2001		
Residence: City	Witten OEX	State		Country	Germany	Citizenship	Germany ✓
Post Office Address	Sauerbruchstr. 3a						
Post Office Address							
City	58453 Witten	State		Zip		Country	Germany
Applicant Authority							

4-00

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Michael	Middle Initial		Family Name	MEINDERS	Suffix e.g. Jr.	
Inventor's Signature	Michael Meinders			Date	August 1, 2001		
Residence: City	Krefeld OEX	State		Country	Germany	Citizenship	Germany
Post Office Address	Am Eickerhof 11						
Post Office Address							
City	47800 Krefeld	State		Zip		Country	Germany
Applicant Authority							

5-00

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Peter	Middle Initial		Family Name	WUELKNITZ	Suffix e.g. Jr.	
Inventor's Signature	Peter Wuelknitz			Date	August 1, 2001		
Residence: City	Leichlingen OEX	State		Country	Germany	Citizenship	Germany
Post Office Address	Im Erlengrund 9						
Post Office Address							
City	42799 Leichlingen	State		Zip		Country	Germany
Applicant Authority							

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DECLARATION

ADDITIONAL INVENTOR(S) Supplemental Sheet

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Rolf	Middle Initial		Family Name	HEMPELMANN	Suffix e.g. Jr.	
Inventor's Signature	Rolf Hempel				Date	August 1, 2001	
Residence: City	St. Ingbert OEX	State		Country	Germany	Citizenship	Germany
Post Office Address	St. Herblainer Str. 11						
Post Office Address							
City	66386 St. Ingbert	State		Zip		Country	Germany	Applicant Authority	

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Marcel	Middle Initial		Family Name	ROTH	Suffix e.g. Jr.	
Inventor's Signature					Date		
Residence: City	Duesseldorf	State		Country	Germany	Citizenship	Germany
Post Office Address	Weststrasse 17						
Post Office Address							
City	40597 Duesseldorf	State		Zip		Country	Germany	Applicant Authority	

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name		Suffix e.g. Jr.	
Inventor's Signature					Date		
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		Zip		Country		Applicant Authority	

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name		Suffix e.g. Jr.	
Inventor's Signature					Date		
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		Zip		Country		Applicant Authority	

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DECLARATION

ADDITIONAL INVENTOR(S) Supplemental Sheet

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Rolf	Middle Initial		Family Name	HEMPELMANN	Suffix e.g. Jr.	
Inventor's Signature					Date		
Residence: City	St. Ingbert	State		Country	Germany	Citizenship	Germany
Post Office Address	St. Herblainer Str. 11						
Post Office Address							
City	66386 St. Ingbert	State		Zip		Country	Germany	Applicant Authority	

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name	Marcel	Middle Initial		Family Name	ROTH	Suffix e.g. Jr.	
Inventor's Signature	Marcel Roth				Date	August 1, 2001	
Residence: City	Duesseldorf DEX	State		Country	Germany	Citizenship	Germany
Post Office Address	Weststrasse 17						
Post Office Address							
City	40597 Duesseldorf	State		Zip		Country	Germany	Applicant Authority	

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name		Suffix e.g. Jr.	
Inventor's Signature					Date		
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		Zip		Country		Applicant Authority	

Name of Additional Joint Inventor, if any: ☐ A petition has been filed for this unsigned inventor

Given Name		Middle Initial		Family Name		Suffix e.g. Jr.	
Inventor's Signature					Date		
Residence: City		State		Country		Citizenship	
Post Office Address							
Post Office Address							
City		State		Zip		Country		Applicant Authority	

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